

Europäisches **Patentamt**

European **Patent Office** Office européen des brevets

02 09. 2004

Bescheinigung

Certificate

Attestalion

13 OCT 2004 **REC'D**

WIPO

Die angehefteten Unterlagen stimmen mit der ursprünglich eingereichten Fassung der auf dem nächsten Blatt bezeichneten europäischen Patentanmeldung überein.

The attached documents are exact copies of the European patent application described on the following page, as originally filed.

Les documents fixés à cette attestation sont conformes à la version initialement déposée de la demande de brevet européen spécifiée à la page suivante.

Patentanmeldung Nr.

Patent application No. Demande de brevet n°

03102707.1

SUBMITTED OR TRANSMITTED IN COMPLIANCE WITH RULE 17.1(a) OR (b)

Der Präsident des Europäischen Patentamts;

For the President of the European Patent Office

Le Président de l'Office européen des brevets p.o.

R C van Dijk

THAILABLE COPY



European Patent Office Office européen des brevets



Anmeldung Nr:

Application no.: 03102707.1

Demande no:

Anmeldetag:

Date of filing: 05.09.03

Date de dépôt:

Anmelder/Applicant(s)/Demandeur(s):

Ciba Specialty Chemicals Holding Inc. Klybeckstrasse 141 4002 Basel SUISSE

Bezeichnung der Erfindung/Title of the invention/Titre de l'invention: (Falls die Bezeichnung der Erfindung nicht angegeben ist, siehe Beschreibung. If no title is shown please refer to the description. Si aucun titre n'est indiqué se referer à la description.)

Electroluminescent device

In Anspruch genommene Prioriät(en) / Priority(ies) claimed /Priorité(s) revendiquée(s)
Staat/Tag/Aktenzeichen/State/Date/File no./Pays/Date/Numéro de dépôt:

Internationale Patentklassifikation/International Patent Classification/Classification internationale des brevets:

H05B33/00

Am Anmeldetag benannte Vertragstaaten/Contracting states designated at date of filing/Etats contractants désignées lors du dépôt:

AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HU IE IT LI LU MC NL PT RO SE SI SK TR

ELECTROLUMINESCENT DEVICE

The present invention relates to organo-electroluminescent (EL) devices, in particular EL devices that comprise durable, blue-emitting organo-electrouminescent layers. The organo-electroluminescent layers comprise certain pyridine compounds.

The present invention is aimed at an electroluminescent device comprising an organic lightemitting layer that contains at least one blue-emitting pyridine compound.

A. Kurfürst et al., Collect. Czech. Chem. Commun. 54 (1989) 462-472 relates to photoluminescent 2,4,6-triarylpyridines of the following formula:

	m	n
1a	1	1
1b	1	2
1c	1	3
1d	2	2
1e	2	3
1f	3	3

and X is C or N.

EP-A-1,202,608 discloses EL devices comprising a carbazole compound of formula

which constitutes the hole transporting layer.

JP2002324678 relates to light emitting elements comprising at least one kind of compound of

Ar¹¹, Ar²¹ and Ar³¹ denote arylene groups, Ar¹², Ar²² and Ar³² denote substituents or hydrogen atoms, wherein at least one of Ar¹¹, Ar²¹, Ar³¹, Ar¹², Ar²² and Ar³² is either a condensed ring aryl structure or a condensed ring heteroaryl structure; Ar denotes an arylene group or a heteroarylene group; and at least one amine derivative having a condensed ring group with two or more rings are contained in a luminous layer. As examples of compounds of the above formula, wherein Ar denotes a heteroarylene group the following two compounds are explicitly mentioned:

It is the object of the present invention to provide a light emitting element with excellent light emitting characteristics and durability.

Accordingly the present invention relates to an electroluminescent device comprising an anode, a cathode and one or a plurality of organic compound layers sandwiched therebetween, in which said organic compound layers comprise a pyridine compound of formula

especially a group of formula
$$R^{41}R^{42}$$
, $R^{44}R^{44'}$, $R^{46}R^{46'}$, or

 R^{11} , R^{12} , R^{12} , R^{13} , R^{13} , R^{15} , R^{16} , R^{16} , R^{16} , R^{17} , R^{17} , R^{41} , R^{41} , R^{42} , R^{42} , R^{44} , R^{44} , R^{45} , R^{46} , R^{46} , R^{46} , R^{46} , R^{47} and R^{47} are independently of each other H, E, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; C_7 - C_{18} aralkyl; or C_7 - C_{18} aralkyl which is substituted by E; or

R^{11'} and R¹², R^{12'} and R¹³, R^{15'} and R¹⁶, R^{16'} and R¹⁷, R^{44'} and R⁴⁶ and/or R^{45'} and R⁴⁷ are each a divalent group L¹ selected from an oxygen atom, an sulfur atom, >CR¹⁸R¹⁹ >SiR¹⁸R¹⁹, or

 R^{18} and R^{19} are independently of each other C_1 - C_{18} alkyl; C_1 - C_{18} alkoxy, C_6 - C_{18} aryl; C_7 - C_{18} aralkyl;

 R^{11} and $R^{11'}$, R^{12} and $R^{12'}$, R^{13} and $R^{13'}$, $R^{13'}$ and R^{14} , R^{14} and R^{15} , R^{15} and $R^{15'}$, R^{16} and $R^{16'}$, $R^{17'}$ and $R^{17'}$, R^{41} and $R^{41'}$, R^{42} and $R^{42'}$, $R^{42'}$ and R^{43} , $R^{41'}$ and R^{43} , R^{44} and $R^{44'}$, R^{45} and $R^{45'}$, $R^{46'}$ and $R^{46'}$, $R^{47'}$ and $R^{47'}$, $R^{46'}$ and R^{48} and $R^{48'}$ and $R^{48'}$ are each a divalent group

 R^{30} , R^{31} , R^{32} , R^{33} , R^{49} and R^{50} are independently of each other H, C₁-C₁₈alkyl; C₁-C₁₈alkyl, which is substituted by E and/or interrupted by D; E; C₆-C₁₈aryl; C₆-C₁₈aryl, which is substituted by E;

R¹⁴ is H, C₂-C₃₀heteroaryl, C₆-C₃₀aryl, or C₆-C₃₀aryl which is substituted by E, C₁-C₁₈alkyl; or

$$- R^{21}$$

$$- R^{22}$$

$$R^{23}$$

C₁-C₁₈alkyl which is substituted by E and/or interrupted by D; especially

 R^{26} and R^{27} are independently of each other H, E, C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; E; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E;

 R^{43} and R^{48} are independently of each other H, E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl, which is substituted by E and/or interrupted by D; C_2 - C_{30} heteroaryl; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E;

D is -CO-; -COO-; -S-; -SO-; -SO₂-; -O-; -NR⁵-; SiR¹¹R¹²-; -POR⁵-; -CR⁹=CR¹⁰-; or -C=C-;

E is -OR⁵; -SR⁵; -NR⁵R⁶; -COR⁸; -COOR⁷; -CONR⁵R⁶; -CN; or halogen, especially F or CI; wherein R⁵ and R⁶ are independently of each other C₆-C₁₈aryl; C₆-C₁₈aryl which is substituted by C₁-C₁₈alkyl, C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-; or

R⁵ and R⁶ together form a five or six membered ring, in particular

 R^7 is C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by -O-;

R⁸ is C₇-C₁₂alkylaryl; C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-;

 R^9 and R^{10} are independently of each other H, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by --O-; and

 R^{11} and R^{12} are independently of each other C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by --O-; with the proviso that

In general, the pyridine compound or compounds emit light below about 520 nm, in particular between about 310 nm and about 520 nm.

The pyridine compound or compounds have a NTSC coordinate of between about (0.12, 0.05) and about (0.16, 0.10), preferably a NTSC coordinate of about (0.14, 0.08).

The pyridine compound or compounds have a melting point above about 150°C, preferably above about 200°C and most preferred above about 250°C.

To obtain organic layers of this invention with the proper T_g , or glass transition temperature, it is advantageous that the present organic compounds have a melting point greater than about 150°C, for example greater than about 200°C, for example greater than about 250°C, for instance greater than about 300°C.

The electroluminescent devices of the present invention are otherwise designed as is known in the art, for example as described in U.S. Pat. Nos. 5,518,824, 6,225,467, 6,280,859, 5,629,389, 5,486,406, 5,104,740, 5,116,708 and 6,057,048, the relevant disclosures of which are hereby incorporated by reference.

For example, organic EL devices contain one or more layers such as:

substrate; base electrode; hole-injecting layer; hole transporting layer; emitter layer; electron-transporting layer; electron-injecting layer; top electrode; contacts and encapsulation.

This structure is a general case and may have additional layers or may be simplified by omitting layers so that one layer performs a plurality of tasks. For instance, the simplest organic EL device consists of two electrodes which sandwich an organic layer that performs all functions, including the function of light emission.

A preferred EL device comprises in this order:

- (a) an anode,
- (b) a hole injecting layer and/or a hole transporting layer,
- (c) a light-emitting layer,
- (d) optionally an electron transporting layer and
- (e) a cathode.

In particular, the present organic compounds function as light emitters and are contained in the light emission layer or form the light-emitting layer.

The light emitting compounds of this invention exhibit intense fluorescence in the solid state and have excellent electric-field-applied light emission characteristics. Further, the light emitting compounds of this invention are excellent in the injection of holes from a metal electrode and the transportation of holes; as well as being excellent in the injection of electrons from a metal electrode and the transportation of electrons. They are effectively used as light emitting materials and may be used in combination with other hole transporting materials, other electron transporting materials or other dopants.

The organic compounds of the present invention form uniform thin films. The light emitting layers may therefore be formed of the present organic compounds alone.

Alternatively, the light-emitting layer may contain a known light-emitting material, a known dopant, a known hole transporting material or a known electron transporting material as required. In the organic EL device, a decrease in the brightness and life caused by quenching can be prevented by forming it as a multi-layered structure. The light-emitting material, a dopant, a hole-injecting material and an electron-injecting material may be used in combination as required. Further, a dopant can improve the light emission brightness and the light emission efficiency, and can attain the red or blue light emission. Further, each of the hole transporting zone, the light-emitting layer and the electron transporting zone may have the layer structure of at least two layers. In the hole transporting zone in this case, a layer to which holes are injected from an electrode is called "hole-injecting layer", and a layer which receives holes from the hole-injecting layer and transport the holes to a light-emitting layer is called "hole transporting layer". In the electron transporting zone, a layer to which electrons are injected from an electrode is called "electron-injecting layer", and a layer which receives electrons from the electron-injecting layer and transports the electrons to a light-emitting layer is called "electron transporting layer". These layers are selected and used depending upon factors such as the energy level and heat resistance of materials and adhesion to an organic layer or metal electrode.

The light-emitting material or the dopant which may be used in the light-emitting layer together with the organic compounds of the present invention includes for example anthracene, naphthalene, phenanthrene, pyrene, tetracene, coronene, chrysene, fluorescein, perylene, phthaloperylene, naphthaloperylene, perinone, phthaoperinone, diphenylbutadiene, tetraphenylbutadiene, coumarine, naphthaloperinone, oxadiazole. aldazine, bisbenzoxazoline, bisstyryl, pyrazine, cyclopentadiene, quinoline metal complex, aminoquinoline metal complex, benzoquinoline metal complex, imine, diphenylethylene, vinyl anthracene, diaminocarbazole, pyran, thiopyran, polymethine, merocyanine, an imidazolechelated oxynoid compound, quinacridone, rubrene, and fluorescent dyestuffs for a dyestuff laser or for brightening.

The pyridine compounds of the present invention and the above compound or compounds that can be used in a light-emitting layer may be used in any mixing ratio for forming a light-emitting layer. That is, the organic compounds of the present invention may provide a main component for forming a light-emitting layer, or they may be a doping material in another main material, depending upon a combination of the above compounds with the organic compounds of the present invention.

The hole-injecting material is selected from compounds which are capable of transporting holes, are capable of receiving holes from the anode, have an excellent effect of injecting holes to a light-emitting layer or a light-emitting material, prevent the movement of excitons

generated in a light-emitting layer to an electron-injecting zone or an electron-injecting material and have the excellent capability of forming a thin film. Suitable hole-injecting materials include for example a phthalocyanine derivative, a naphthalocyanine derivative, a porphyrin derivative, oxazole, oxadiazole, triazole, imidazole, imidazolone, imidazolthione, pyrazolone, tetrahydroimidazole, oxazole, oxadiazole, hydrazone, acylhydrazone, polyarylalkane, stilbene, butadiene, benzidine type triphenylamine, styrylamine type triphenylamine, diamine type triphenylamine, derivatives of these, and polymer materials such as polyvinylcarbazole, polysilane and an electroconducting polymer.

In the organic EL device of the present invention, the hole-injecting material which is more effective is an aromatic tertiary amine derivative or a phthalocyanine derivative. Although not specially limited, specific examples of the tertiary amine derivative include triphenylamine, tritolylamine, tolyldiphenylamine, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1-biphenyl-4,4'-N,N,N',N'-tetra(4-methylphenyl)-1,1'-phenyl-4,4'-diamine, diamine, N,N,N',N'-tetra(4methylphenyl)-1,1'-biphenyl-4,4'-diamine, N,N'-diphenyl-N,N'-di(1-naphthyl)-1,1'-biphenyl-4,4'-diamine, N,N'-di(methylphenyl)-N,N'-di(4-n-butylphenyl)-phenanthrene-9,10- diamine, 4"-tris(3-methylphenyl)-N-phenylamino)triphenylamine, 4,4', tolylaminophenyl)cyclohexane, and oligomers or polymers having aromatic tertiary amine structures of these.

Although not specially limited, specific examples of the phthalocyanine (Pc) derivative include phthalocyanine derivatives or naphthalocyanine derivatives such as H₂Pc, CuPc, CoPc, NiPc, ZnPc, PdPc, FePc, MnPc, ClAlPc, ClGaPc, ClInPc, ClSnPc, Cl₂SiPc, (HO)AlPc, (HO)GaPc, VOPc, TiOPc, MoOPc, and GaPc-O-GaPc.

The hole transporting layer can reduce the driving voltage of the device and improve the confinement of the injected charge recombination within the pyridine light emitting layer. Any conventional suitable aromatic amine hole transporting materials described for the hole-injecting layer may be selected for forming this layer.

A preferred class of hole transporting materials is comprised of 4,4'-bis(9-carbazolyl)-1,1'-biphenyl compounds of the formula

wherein R^{61} and R^{62} is a hydrogen atom or an C_1 - C_3 alkyl group; R^{63} through R^{66} are substituents independently selected from the group consisting of hydrogen, a C_1 - C_6 alkyl

group, a C_1 - C_6 alkoxy group, a halogen atom, a dialkylamino group, a C_6 - C_{30} aryl group, and the like. Illustrative examples of 4,4'-bis(9-carbazolyl)-1,1'-biphenyl compounds include 4,4'-bis(9-carbazolyl)-1,1'-biphenyl, and the like. The electron transporting layer is not necessarily required for the present device, but is optionally and preferably used for the primary purpose of improving the electron injection characteristics of the EL devices and the emission uniformity. Illustrative examples of electron transporting compounds, which can be utilized in this layer, include the metal chelates of 8-hydroxyquinoline as disclosed in U.S. Pat. Nos. 4,539,507, 5,151,629, and 5,150,006, the disclosures of which are totally incorporated herein by reference.

Although not specially limited, specific examples of the metal complex compound include lithium 8-hydroxyquinolinate, zinc bis(8-hydroxyquinolinate), copper bis(8hydroxyquinolinate), manganese bis(8-hydroxyquinolinate), aluminum tris(8hydroxyguinolinate), aluminum tris(2-methyl-8-hydroxyguinolinate), gallium tris(8hydroxyquinolinate),---beryllium---bis(10-hydroxybenzo[h]quinolinate),---zinc---bis(10hydroxybenzo[h]quinolinate), chlorogallium bis(2-methyl-8-quinolinate), gallium bis(2-methyl-8-quinolinate)(o-cresolate), aluminum bis(2-methyl-8-quinolinate)(1-naphtholate), gallium bis(2-methyl-8-quinolinate)(2-naphtholate), gallium bis(2-methyl-8-quinolinate)phenolate, zinc bis(o-(2-benzooxazolyl)phenolate), zinc bis(o-(2-benzothiazolyl)phenolate) and zinc bis(o-(2benzotrizolyl)phenolate). The nitrogen-containing five-membered derivative is preferably an oxazole, thiazole, thiadiazole, or triazole derivative. Although not specially limited, specific examples of the above nitrogen-containing five-membered derivative include 2,5-bis(1phenyl)-1,3,4-oxazole, 1,4-bis(2-(4-methyl-5-phenyloxazolyi)benzene, 2,5-bis(1-phenyl)-1,3,4-thiazole, 2,5-bis(1-phenyl)-1,3,4-oxadiazole, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)1,3,4oxadiazole, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole, 1,4-bis[2-(5-phenyloxadiazolyl)]benzene, 1,4-bis[2-(5-phenyloxadiazolyl)-4-tert-butylbenzene], 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-thiadiazole, 2,5-bis(1-naphthyl)-1,3,4-thiadiazole, 1,4-bis[2-(5-phenylthiazolyl)]benzene, 2-(4'-tert-butylphenyl)-5-(4"-biphenyl)-1,3,4-triazole, 2,5-bis(1-naphthyl)-1,3,4-triazole 1,4-bis[2-(5-phenyltriazolyl)]benzene. Another class of electron transport materials are oxadiazole metal chelates, such as bis[2-(2-hydroxyphenyl)-5-phenyl-1,3.4oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato]beryllium; bis[2-(2hydroxyphenyl)-5-(1-naphthyl)-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-(1naphthyl)-1,3,4-oxadiazolato]beryllium; bis[5-biphenyl-2-(2-hydroxyphenyl)-1,3,4oxadiazolato]zinc; bis[5-biphenyl-2-(2-hydroxyphenyl)-1,3,4-oxadiazolato]beryllium; bis(2hydroxyphenyl)-5-phenyl-1,3,4-oxadiazolato]lithium; bis[2-(2-hydroxyphenyl)-5-p-tolyl-1,3,4oxadiazolato]zinc; bis 2-(2-hydroxyphenyl)-5-p-tolyl-1,3,4-oxadiazolato]beryllium; bis[5-(ptert-butylphenyl)-2-(2-hydroxyphenyl)-1,3,4-oxadiazolato]zinc; bis[5-(p-tert-butylphenyl)-2-(2hydroxyphenyl)-1,3,4-oxadiazolato]beryllium; bis[2-(2-hydroxyphenyl)-5-(3-fluorophenyl)-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-(4-fluorophenyl)-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-(4-fluorophenyl)-1,3,4-oxadiazolato]beryllium; bis[5-(4chlorophenyl)-2-(2-hydroxyphenyl)-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxy phenyl)-5-(4methoxyphenyl)-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxy-4-methylphenyl)-5-phenyl-1,3,4oxadiazolato]zinc; bis[2-.alpha.-(2-hydroxynaphthyl)-5-phenyl-1,3,4-oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-p-pyridyl-1,3,4-oxadiazolatojzinc; bis[2-(2-hydroxyphenyl)-5-p-pyridyl-1,3,4-oxadiazolato]beryllium; bis[2-(2-hydroxyphenyl)-5-(2-thiophenyl)-1,3,4oxadiazolato]zinc; bis[2-(2-hydroxyphenyl)-5-phenyl-1,3,4-thiadiazolato]zinc; hydroxyphenyl)-5-phenyl-1,3,4-thiadiazolato]beryllium; bis[2-(2-hydroxyphenyl)-5-(1naphthyl)-1,3,4-thiadiazolatojzinc; and bis[2-(2-hydroxyphenyl)-5-(1-naphthyl)-1,3,4thiadiazolato]beryllium, and the like.

In the organic EL device of the present invention, the light-emitting layer may contain, in addition to the light-emitting organic material of the present invention, at least one of other light-emitting material, other dopant, other hole-injecting material and other electron-injecting material. For improving the organic EL device of the present invention in the stability against temperature, humidity and ambient atmosphere, a protective layer may be formed on the surface of the device, or the device as a whole may be sealed with a silicone oil, or the like.

The electrically conductive material used for the anode of the organic EL device is suitably selected from those materials having a work function of greater than 4 eV. The electrically conductive material includes carbon, aluminum, vanadium, iron, cobalt, nickel, tungsten, silver, gold, platinum, palladium, alloys of these, metal oxides such as tin oxide and indium oxide used for ITO substrates or NESA substrates, and organic electroconducting polymers such as polythiophene and polypyrrole.

The electrically conductive material used for the cathode is suitably selected from those having a work function of smaller than 4 eV. The electrically conductive material includes magnesium, calcium, tin, lead, titanium, yttrium, lithium, ruthenium, manganese, aluminum and alloys of these, while the electrically conductive material shall not be limited to these. Examples of the alloys include magnesium/silver, magnesium/indium and lithium/aluminum, while the alloys shall not be limited to these. Each of the anode and the cathode may have a layer structure formed of two layers or more as required.

For the effective light emission of the organic EL device, at least one of the electrodes is desirably sufficiently transparent in the light emission wavelength region of the device. Further, the substrate is desirably transparent as well. The transparent electrode is produced from the above electrically conductive material by a deposition method or a sputtering method such that a predetermined light transmittance is secured. The electrode

on the light emission surface side has for instance a light transmittance of at least 10%. The substrate is not specially limited so long as it has adequate mechanical and thermal strength and has transparency. For example, it is selected from glass substrates and substrates of transparent resins such as a polyethylene substrate, a polyethylene terephthalate substrate, a polyether sulfone substrate and a polypropylene substrate.

In the organic EL device of the present invention, each layer can be formed by any one of dry film forming methods such as a vacuum deposition method, a sputtering method, a plasma method and an ion plating method and wet film forming methods such as a spin coating method, a dipping method and a flow coating method. The thickness of each layer is not specially limited, while each layer is required to have a proper thickness. When the layer thickness is too large, inefficiently, a high voltage is required to achieve predetermined emission of light. When the layer thickness is too small, the layer is liable to have a pinhole, etc., so that sufficient light emission brightness is hard to obtain when an electric field is applied. The thickness of each layer is for example in the range of from about 5 nm to about 10 μ m, for instance about 10 nm to about 0.2 μ m.

In the wet film forming method, a material for forming an intended layer is dissolved or dispersed in a proper solvent such as ethanol, chloroform, tetrahydrofuran and dioxane, and a thin film is formed from the solution or dispersion. The solvent shall not be limited to the above solvents. For improving the film formability and preventing the occurrence of pinholes in any layer, the above solution or dispersion for forming the layer may contain a proper resin and a proper additive. The resin that can be used includes insulating resins such as polystyrene, polycarbonate, polyarylate, polyester, polyamide, polyurethane, polysulfone, polymethyl methacrylate, polymethyl acrylate and cellulose, copolymers of these, photoconductive resins such as poly-N-vinylcarbozole and polysilane, and electroconducting polymers such as polythiophene and polypyrrole. The above additive includes an antioxidant, an ultraviolet absorbent and a plasticizer.

When the light-emitting organic material of the present invention is used in a light-emitting layer of an organic EL device, an organic EL device can be improved in organic EL device characteristics such as light emission efficiency and maximum light emission brightness. Further, the organic EL device of the present invention is remarkably stable against heat and electric current and gives a usable light emission brightness at a low actuation voltage. The problematic deterioration of conventional devices can be remarkably decreased.

The organic EL device of the present invention has significant industrial values since it can be adapted for a flat panel display of an on-wall television set, a flat light-emitting device, a light source for a copying machine or a printer, a light source for a liquid crystal display or counter, a display signboard and a signal light.

The material of the present invention can be used in the fields of an organic EL device, an electrophotographic photoreceptor, a photoelectric converter, a solar cell, an image sensor, dye lasers and the like.

The pyridine compounds of formula I are novel. Hence, a further subject of the present invention is directed to pyridine compounds of formula

especially a group of formula

 R^{11} , R^{12} , R^{12} , R^{12} , R^{13} , R^{13} , R^{15} , R^{15} , R^{16} , R^{16} , R^{17} , R^{17} , R^{41} , R^{41} , R^{42} , R^{42} , R^{44} , R^{44} , R^{45} , R^{45} , R^{46} , R^{46} , R^{46} , R^{47} and R^{47} are independently of each other H, E, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; C_7 - C_{18} aralkyl; or C_7 - C_{18} aralkyl which is substituted by E; or

 $R^{11'}$ and R^{12} , $R^{12'}$ and R^{13} , $R^{15'}$ and R^{16} , $R^{16'}$ and R^{17} , $R^{44'}$ and R^{46} and/or $R^{45'}$ and R^{47} are each a divalent group L^1 selected from an oxygen atom, an sulfur atom, $>CR^{18}R^{19} > SiR^{18}R^{19}$, or

 R^{18} and R^{19} are independently of each other C_1 - C_{18} alkyl; C_1 - C_{18} alkoxy, C_6 - C_{18} aryl; C_7 - C_{18} aralkyl;

 R^{11} and $R^{11'}$, R^{12} and $R^{12'}$, R^{13} and $R^{13'}$, $R^{13'}$ and R^{14} , R^{14} and R^{15} , R^{15} and R^{15} , R^{16} and $R^{16'}$, $R^{17'}$ and R^{17} , R^{41} and $R^{41'}$, R^{42} and $R^{42'}$, $R^{42'}$ and R^{43} , $R^{41'}$ and R^{43} , R^{44} and $R^{44'}$, R^{45} and $R^{45'}$, R^{46}

and R⁴⁶, R⁴⁷ and R⁴⁷, R⁴⁶ and R⁴⁸ and/or R⁴⁷ and R⁴⁸ are each a divalent group

$$R^{32}$$
 R^{33}
 R^{30}
, wherein

 R^{30} , R^{31} , R^{32} , R^{33} , R^{49} and R^{50} are independently of each other H, C₁-C₁₈alkyl; C₁-C₁₈alkyl, which is substituted by E and/or interrupted by D; E; C₆-C₁₈aryl; C₆-C₁₈aryl, which is substituted by E;

R¹⁴ is H, C₂-C₃₀heteroaryl, C₆-C₃₀aryl, or C₆-C₃₀aryl which is substituted by E, C₁-C₁₈alkyl; or

$$- \bigvee_{R^{23}}^{R^{21}} R^{22}$$

C1-C18alkyl which is substituted by E and/or interrupted by D; especially

 R^{28} and R^{27} are independently of each other H, E, C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; E; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E;

 R^{43} and R^{48} are independently of each other H, E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl, which is substituted by E and/or interrupted by D; C_2 - C_{30} heteroaryl; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E;

D is -CO-; -COO-; -S-; -SO-; -SO₂-; -O-; -NR⁵-; SiR¹¹R¹²-; -POR⁵-; -CR⁹=CR¹⁰-; or -C=C-;

E is $-OR^5$; $-SR^5$; $-NR^5R^6$; $-COR^8$; $-COOR^7$; $-CONR^5R^6$; -CN; or halogen, especially F, or CI; wherein R^5 and R^6 are independently of each other C_6-C_{18} aryl; C_6-C_{18} aryl which is substituted by C_1-C_{18} alkyl, C_1-C_{18} alkyl; or C_1-C_{18} alkyl which is interrupted by $-C_1$; or

R⁵ and R⁶ together form a five or six membered ring, in particular

$$-\mathbf{N}$$

 R^7 is C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by -O-;

R⁸ is C₇-C₁₂alkylaryl; C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-;

R9 and R10 are independently of each other H, C6-C18 aryl; C6-C18 aryl which is substituted by C₁-C₁₈alkyl, C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-; and

R¹¹ and R¹² are independently of each other C₆-C₁₈aryl; C₆-C₁₈aryl which is substituted by C₁-C₁₈alkyl, C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-; with the proviso that

compounds of formula I, wherein Y is

W is preferably a group of formula

R¹³, R¹³, R¹⁵ and R¹⁵ are H and R²⁰ is H, especially

R¹³ and R¹⁵ are H, R¹³ and R¹⁵ are independently of each other H, C₁-C₈alkyl, or C₁-C₈alkoxy, and R²⁰ is H, C₁-C₈alkyl, or C₁-C₈alkoxy; or

$$R^{32}$$
 R^{31} R^{33} R^{30} R^{33} R^{30} R^{31} and R^{15} are H, and R^{13} and R^{20} are

R²⁰. R¹⁵ and R¹⁵ are H. and R¹³ and R¹³ are wherein

R³⁰, R³¹, R³² and R³³ are H, C₁-C₈alkyl, or C₁-C₈alkoxy, and X and Y are as defined above.

According to the present invention at least one of the groups W, X and Y, preferably two of the groups W, X and Y, most preferred all three groups W, X and Y are a group of formula

especially H, C₁-C₈alkyl, C₁-C₈alkoxy, or phenyl.

Accordingly, in one preferred embodiment of the present invention the pyridine compound is a compound of formula I, wherein W and Y or W and X (= X and Y) are independently of

R¹¹, R¹¹, R¹², R¹², R¹³, R¹³, R¹⁵, R¹⁵, R¹⁶, R¹⁶, R¹⁶, R¹⁷ and R¹⁷, R⁴¹, R⁴¹, R⁴², R⁴², R⁴⁴, R⁴⁴, R⁴⁵, R⁴⁵, R⁴⁶, R⁴⁶, R⁴⁶, R⁴⁷, and R⁴⁷ as well as R¹⁴, R⁴³, and R⁴⁸ are preferably independently of each other H, E; or C₁-C₈alkyl, especially H, C₁-C₄alkyl, C₁-C₄alkoxy, or phenyl; wherein E is -OR⁵; -SR⁵; -NR⁵R⁶; -COR⁸; -COOR⁷; -CONR⁵R⁶; -CN; -OCOOR⁷; or halogen, especially F; wherein R⁵ and R⁶ are independently of each other C₆-C₁₂ary, or C₁-C₈alkyl;

R7 is C7-C12 alkylaryl, or C1-C8alkyl; and

 R^8 is C_6 - C_{12} aryl; or C_1 - C_8 alkyl, or

 R^{11} and $R^{11'}$, R^{12} and $R^{12'}$, R^{13} and $R^{13'}$, $R^{13'}$ and R^{14} , R^{41} and $R^{41'}$, $R^{41'}$ and R^{43} , R^{44} and $R^{44'}$,

 R^{46} and R^{46} , $R^{46'}$ and R^{48} and/or $R^{47'}$ and R^{48} are each a divalent group

In one more preferred embodiment of the present invention W, X and Y are independently of each other a group of formula

 R^{11} , R^{12} , R^{12} , R^{12} , R^{13} , R^{13} , R^{15} , R^{15} , R^{16} , R^{16} , R^{17} and R^{17} are independently of each other H, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by E; E, C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E; and R^{14} , R^{18} and R^{19} are as defined above, or

W is a group of the formula -W1-W2-W3,

X is a group of the formula -X1-X2-X3 and

Y is a group of the formula $-Y^1-Y^2-Y^3$, wherein W^1 , W^2 , X^1 , X^2 , Y^1 and Y^2 are independently of each other a group of formula

and W^3 , X^3 and Y^3 are independently of each

$$-$$
R¹⁴, $-$ R¹⁴, or $-$ R¹⁴

other a group of formula defined above.

wherein R¹⁴ is as

W, X and Y can be different, but have preferably the same meaning.

Pyridine compounds of formula I are preferred, wherein R^{11} , $R^{11'}$, R^{12} , R^{12} , R^{13} , $R^{13'}$, R^{15} , $R^{15'}$, R^{16} , $R^{16'}$, R^{17} and $R^{17'}$, $R^{41'}$, R^{42} , R^{42} , R^{44} , $R^{44'}$, R^{45} , $R^{45'}$, R^{46} , $R^{46'}$, R^{47} , and $R^{47'}$ are independently of each other H, E; or C_1 - C_8 alkyl; wherein

E is $-OR^5$; $-SR^5$; $-NR^5R^6$; $-COR^8$; $-COR^7$; $-CONR^5R^6$; -CN; $-OCOOR^7$; or halogen; wherein R^5 and R^6 are independently of each other C_6-C_{12} aryl; or C_1-C_8 alkyl;

R⁷ is C₇-C₁₂ alkylaryl, or C₁-C₈alkyl; and

 R^8 is C_6 - C_{12} aryl, or C_1 - C_8 alkyl.

Especially preferred are pyridine compounds of formula I, wherein

W, X and Y are independently of each other a group of formula

, wherein R^{13} , $\mathsf{R}^{13'}$, R^{15} and $\mathsf{R}^{15'}$ are H and R^{20} is H,

 R^{13} and R^{15} are H, $R^{13'}$ and $R^{15'}$ are independently of each other H, C_1 - C_8 alkyl, or C_1 - C_8 alkoxy; or

$$R^{32}$$
 R^{31} R^{33} R^{30} R^{13} , R^{15} and R^{15} are H, and R^{13} and R^{20} are R^{32} , or

$$R^{32}$$
 R^{31} R^{30}

 R^{20} , R^{15} and R^{15} are H, and R^{13} and R^{13} are

, wherein

 $\mathsf{R}^{30},\,\mathsf{R}^{31},\,\mathsf{R}^{32}$ and R^{33} are H, C1-C8alkyl, or C1-C8alkoxy; or wherein

W, X and Y are independently of each other a group of formula

wherein R^{18} and R^{19} are independently of each other C_1 - C_8 alkyl.

Specific examples of preferred pyridine compounds are:

The present pyridine compounds can be prepared according to or analogous to known procedures. The pyridine compounds of the present invention of the formula:

can, for example, be prepared according to a process, which comprises reacting a derivative of formula

wherein R^{100} stands for halogen such as chloro or bromo, preferably bromo (cf. F. Kröhnke, Synthesis 1976, 1), or E having the meaning of

wherein a is 2 or 3,

with boronic acid derivative

E-Ar.

or - in case R¹⁰⁰ is not halogen -

Hal-Ar,

wherein Hal stands for halogen, preferably for bromo,

wherein_Ar_is_C₁₂-C₃₀-aryl,_which_can_be_substituted,_especially______,
or

in the presence of an allylpalladium catalyst of the μ -halo(triisopropylphosphine)(η^3 -allyl)palladium(II) type (see for example WO99/47474).

Accordingly, unsymmetrical substituted pyridine compounds of the present invention of the formula:

can, for example, be prepared according to a process, which comprises reacting a derivative of formula 1 (cf. F. Kröhnke, Synthesis 1976, 1)

with a boronic acid derivative E-Ar, wherein E is as defined above.

C₁-C₁₈alkyl is a branched or unbranched radical such as for example methyl, ethyl, propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, 2-ethylbutyl, n-pentyl, isopentyl, 1-methylpentyl, 1,3-dimethylbutyl, n-hexyl, 1-methylhexyl, n-heptyl, isoheptyl, 1,1,3,3-tetramethylbutyl, 1-methylheptyl, 3-methylheptyl, n-octyl, 2-ethylhexyl, 1,1,3-trimethylhexyl, 1,1,3,3-tetramethylpentyl, nonyl, decyl, undecyl, 1-methylundecyl, dodecyl, 1,1,3,3,5,5-hexamethylhexyl, tridecyl, tetradecyl, pentadecyl, hexadecyl, heptadecyl, or octadecyl.

C₁-C₁₈Alkoxy radicals are straight-chain or branched alkoxy radicals, e.g. methoxy, ethoxy, n-propoxy, isopropoxy, n-butoxy, sec-butoxy, tert-butoxy, amyloxy, isoamyloxy or tert-amyloxy, heptyloxy, octyloxy, isooctyloxy, nonyloxy, decyloxy, undecyloxy, dodecyloxy, tetradecyloxy, pentadecyloxy, hexadecyloxy, heptadecyloxy and octadecyloxy.

C₂-C₁₈Alkenyl radicals are straight-chain or branched alkenyl radicals, such as e.g. vinyl, allyl, methallyl, isopropenyl, 2-butenyl, 3-butenyl, isobutenyl, n-penta-2,4-dienyl, 3-methyl-but-2-enyl, n-oct-2-enyl, n-dodec-2-enyl, isododecenyl, n-dodec-2-enyl or n-octadec-4-enyl.

C₂₋₂₄Alkynyl is straight-chain or branched and preferably C₂₋₈alkynyl, which may be unsubstituted or substituted, such as, for example, ethynyl, 1-propyn-3-yl, 1-butyn-4-yl, 1-pentyn-5-yl, 2-methyl-3-butyn-2-yl, 1,4-pentadiyn-3-yl, 1,3-pentadiyn-5-yl, 1-hexyn-6-yl, cis-3-methyl-2-penten-4-yn-1-yl, trans-3-methyl-2-penten-4-yn-1-yl, 1,3-hexadiyn-5-yl, 1-octyn-8-yl, 1-nonyn-9-yl, 1-decyn-10-yl or 1-tetracosyn-24-yl,

 C_4 - C_{18} cycloalkyl is preferably C_5 - C_{12} cycloalkyl, such as, for example, cyclopentyl, cyclohexyl, cycloheptyl, cycloactyl, cyclononyl, cyclodecyl, cyclododecyl are most preferred.

The term "aryl group" is typically C_6 - C_{30} aryl, such as phenyl, indenyl, azulenyl, naphthyl, biphenyl, terphenylyl or quadphenylyl, as-indacenyl, s-indacenyl, acenaphthylenyl, phenanthryl, fluoranthenyl, triphenlenyl, chrysenyl, naphthacen, picenyl, perylenyl, pentaphenyl, hexacenyl, pyrenyl, or anthracenyl, preferably phenyl, 1-naphthyl, 2-naphthyl, 9-phenanthryl, 2- or 9-fluorenyl, 3- or 4-biphenyl, which may be unsubstituted or substituted. Examples of C_6 - C_{18} aryl are phenyl, 1-naphthyl, 2-naphthyl, 3- or 4-biphenyl, 9-phenanthryl, 2- or 9-fluorenyl, which may be unsubstituted or substituted.

 C_7 - C_{24} aralkyl radicals are preferably C_7 - C_{18} aralkyl radicals, which may be substituted, such as, for example, benzyl, 2-benzyl-2-propyl, β -phenyl-ethyl, α,α -dimethylbenzyl, ω -phenyl-butyl, ω -phenyl-butyl, ω -phenyl-octadecyl,

ω-phenyl-eicosyl or ω-phenyl-docosyl, preferably C₇-C₁₈aralkyl such as benzyl, β-phenyl-ethyl, α , α -dimethylbenzyl, ω-phenyl-butyl, 2-benzyl-2-propyl, ω,ω-dimethyl-ω-phenyl-butyl, ω-phenyl-dodecyl or ω-phenyl-octadecyl, and particularly preferred C₇-C₁₂aralkyl such as benzyl. 2-benzyl-2-propyl, β-phenyl-ethyl, α,α -dimethylbenzyl, ω -phenyl-butyl, or ω,ω -dimethyl- ω -phenyl-butyl, in which both the aliphatic hydrocarbon group and aromatic hydrocarbon group may be unsubstituted or substituted.

 C_{7} - C_{12} alkylaryl is, for example, a phenyl group substituted with one, two or three C_{1} - C_{6} alkyl groups, such as, for example, 2-, 3-, or 4-methylphenyl, 2-, 3-, or 4-ethylphenyl, 3-, or 4-isopropylphenyl, 3,4-dimethylphenyl, 3,5-dimethylphenyl, or 3,4,5-trimethylphenyl.

The term "heteroaryl group", especially C₂C₃₀heteroaryl, is a ring, wherein nitrogen, oxygen or sulfur are the possible hetero atoms, and is typically an unsaturated heterocyclic radical with five to 18 atoms having at least six conjugated π -electrons such as thienyl, benzo[b]thienyl, dibenzo[b,d]thienyl, thianthrenyl, furyl, furfuryl, 2H-pyranyl, benzofuranyl, isobenzofuranyl, 2H-chromenyl, xanthenyl, dibenzofuranyl, phenoxythienyl, pyrrolyl, imidazolyl, pyrazolyl, pyridyl, bipyridyl, triazinyl, pyrimidinyl, pyrazinyl, 1H-pyrrolizinyl, isoindolyl, pyridazinyl, indolizinyl, isoindolyl, indolyl, 3H- indolyl, phthalazinyl, naphthyridinyl, quinoxalinyl, quinazolinyl, cinnolinyl, indazolyl, purinyl, quinolizinyl, chinolyl, isochinolyl, phthalazinyl, naphthyridinyl, chinoxalinyl, chinazolinyl, cinnolinyl, pteridinyl, carbazolyl, 4aHcarbazolyl, carbolinyl, benzotriazolyl, benzoxazolyl, phenanthridinyl, acridinyl, perimidinyl, isothiazolyl, phenothiazinyl, isoxazolyi, furazanyl phenanthrolinyl, phenazinyl, phenoxazinyl, preferably the above-mentioned mono- or bicyclic heterocyclic radicals, which may be unsubstituted or substituted.

Halogen is fluorine, chlorine, bromine and iodine.

Examples of a five or six membered ring formed by R⁵ and R⁶ are heterocycloalkanes or heterocycloalkenes having from 3 to 5 carbon atoms which can have one additional hetero

Possible substituents of the above-mentioned groups are C₁-C₈alkyl, a hydroxyl group, a mercapto group, C₁-C₈alkoxy, C₁-C₈alkylthio, halogen, halo-C₁-C₈alkyl, a cyano group, an aldehyde group, a ketone group, a carboxyl group, an ester group, a carbamoyl group, an amino group, a nitro group or a silyl group.

As described above, the aforementioned radicals may be substituted by E and/or, if desired, interrupted by D. Interruptions are of course possible only in the case of radicals containing at least 2 carbon atoms connected to one another by single bonds; C₆-C₁₈aryl is not interrupted; interrupted arylalkyl or alkylaryl contains the unit D in the alkyl moiety. C₁-C₁₈alkyl substituted by one or more E and/or interrupted by one or more units D is, for example, (CH₂CH₂O)_n-R^x, where n is a number from the range 1-9 and R^x is H or C₁-C₁₀alkyl or C₂-C₁₀alkanoyl (e.g. CO-CH(C₂H₅)C₄H₉), CH₂-CH(OR^y)-CH₂-O-R^y, where R^y is C₁-C₁₈alkyl, C₅-C₁₂cycloalkyl, phenyl, C₇-C₁₅phenylalkyl, and R^y embraces the same definitions as R^y or is H; C₁-C₈alkylene-COO-R^z, e.g. CH₂COOR_z, CH(CH₃)COOR^z, C(CH₃)₂COOR^z, where R^z is H, C₁-C₁₈alkyl, (CH₂CH₂O)₁₋₉-R^x, and R^x embraces the definitions indicated above; CH₂CH₂-O-CO-CH=CH₂; CH₂CH(OH)CH₂-O-CO-C(CH₃)=CH₂.

The electroluminescent devices may be employed for full color display panels in, for example, mobile phones, televisions and personal computer screens.

The following Examples illustrate the invention. In the Examples and throughout this application, the term light emitting material means the present pyridine compounds.

Examples

Example 1

2,4,6-tris-(p-bromophenyl)-pyridine can be prepared as described in A. Hopff, A. Heer *Chimia* (1959) 13, 105-107.

To 0.300 g (0.550 mmol) 2,4,6-tris-(p-bromophenyl)-pyridine in 20 ml toluol 0.38 g (1.93 mmol) 4-biphenylboronic acid are added. Argon is passed through the reaction mixture. 0.95 g (2.92 mmol) of caesium carbonate in 2 ml water are added dropwise to the reaction mixture. Then 20 mg of the catalyst (WO 99/47474) are added. The reaction mixture is refluxed for 4 h and then the solid is filtered of. The product is crystallized from dimethylfrormamide (yield: 0.31 g (74 %); melting point: 315-317 °C).

Application Example 1

Present compound A1, as light emitting material, 2,5-bis(1-naphthyl)-1,3,4-oxadiazole and a polycarbonate resin in a weight ratio of 5:3:2 are dissolved in tetrahydrofuran, and the solution is spin-coated on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 100 nm. An electrode having a thickness of 150 nm is formed thereon from a magnesium/indium alloy having a magnesium/indium mixing ratio of 10/1, to obtain an organic EL device. The device exhibits light emission with excellent brightness and efficiency at a direct current voltage of 5 V.

Application Example 2

Present compound A1 is vacuum-deposited on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 100 nm. An electrode having a thickness of 100 nm is formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The light-emitting layer is formed by deposition under a vacuum of 10⁶ Torr at a substrate temperature of room

temperature. The device shows emission having an excellent brightness and efficiency at a direct current voltage of 5 V.

Application Example 3

Present compound A1 is dissolved in methylene chloride tetrahydrofuran, and the solution is spin-coated on a cleaned glass substrate with an ITO electrode to form a light-emitting layer having a thickness of 50 nm. Then, aluminum bis(2-methyl-8-quinolinate)(2-naphtolate) is vacuum-deposited to form an electron transporting layer having a thickness of 10 nm, and an electrode having a thickness of 100 nm is formed thereon from a magnesium/aluminum alloy having a magnesium/aluminum mixing ratio of 10/1, to obtain an organic EL device. The light-emitting layer and the electron-injecting layer are formed by deposition under a vacuum of 10⁸ Torr at a substrate temperature of room temperature. The device shows an emission having an excellent brightness and efficiency at a direct current voltage of 5 V.

Application Example 4

One of hole transporting materials (H-1) to (H-6) is vacuum-deposited on a cleaned glass substrate with an ITO electrode, to form a hole transporting layer having a thickness of 30 nm. Then, present compound A1 is vacuum-deposited to form a light-emitting layer having a thickness of 30 nm. Further, one of electron transporting materials (E-1) to (E-6) is vacuum-deposited to form an electron transporting layer having a thickness of 30 nm. An electrode having a thickness of 150 nm is formed thereon from a magnesium/silver alloy having a magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. Each layer is formed under a vacuum of 10⁶ Torr at a substrate temperature of room temperature. All the organic EL devices obtained in these Examples shows high brightness and efficiency.

$$\mathsf{nH_9C_4} \qquad \mathsf{h-6})$$

Application Example 5

On a cleaned glass substrate with an ITO electrode, 4,4',4"-tris(N-(3-methylphenyl)-N-phenylamino)triphenylamine is vacuum-deposited to form a hole-injecting layer having a thickness of 25 nm. Further, a hole transporting material (H-1) is vacuum-deposited to form a hole transporting layer having a thickness of 5 nm. Then, compound A1 as light-emitting material is vacuum-deposited to form a light-emitting layer having a thickness of 20 nm. Further, an electron transporting material (E-1) is vacuum-deposited to form an electron transporting layer having a thickness of 30 nm. Then, an electrode having a thickness of 150 nm is formed thereon from a magnesium/silver alloy having an magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The device shows emission having an outstanding brightness and efficiency at a direct current voltage of 5 V.

Application Example 6

A hole transporting material (H-5) is vacuum-deposited on a cleaned glass substrate with an ITO electrode to form a hole transporting layer having a thickness of 20 nm. Then, compound A1 as light-emitting material is vacuum-deposited to form a light-emitting layer having a thickness of 20 nm. Further, an electron transporting material (E-2) is vacuum-deposited to form a first electron transporting layer having a thickness of 20 nm. Then, an electron transporting material (E-5) is vacuum-deposited to form a second electron transporting layer having a thickness of 10 nm, and an electrode having a thickness of 150 nm is formed thereon from a magnesium/silver alloy having an magnesium/silver mixing ratio of 10/1, to obtain an organic EL device. The device shows light emission having an excellent brightness and efficiency at a direct current voltage of 5 V.

Application Example 7

An organic EL device is prepared in the same manner as in Example 4 except that the light-emitting layer is replaced with a 30 nm thick light-emitting layer formed by vacuum-depositing compound A1 and one of the dopant compounds (D-1) to (D-7) in a weight ratio of 100:1. All

the organic EL devices obtained in these Examples shows high brightness characteristics and gives intended light emission colors.

Application Example 8

On a cleaned glass substrate with an ITO electrode, N,N'-1-naphthyl-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine and 5,10-diphenylanthracene are vacuum-deposited to form a hole-injecting layer. Further, 4,4'-bis(9-carbazolyl)-1,1'-biphenyl is vacuum-deposited to form a hole transporting layer. Then, compound A1 as light-emitting material is vacuum-deposited to form a light-emitting layer. Then, an electrode is formed thereon from a magnesium/silver alloy having an magnesium/silver mixing ratio of 9/1, to obtain an organic EL device. The device shows emission having an outstanding brightness and efficiency at a direct current voltage of 5 V.

The organic EL devices obtained in the Application Examples of the present invention show an excellent light emission brightness and achieved a high light emission efficiency. When the organic EL devices obtained in the above Examples are allowed to continuously emit light at 3 (mA/cm²), all the organic EL devices remain stable. Since the light-emitting materials of the present invention have a very high fluorescence quantum efficiency, the organic EL devices using the light-emitting materials achieved light emission with a high brightness in a low electric current applied region, and when the light-emitting layer

additionally uses a doping material, the organic EL devices are improved in maximum light emission brightness and maximum light emission efficiency. Further, by adding a doping material having a different fluorescent color to the light-emitting material of the present invention, there are obtained light-emitting devices having a different light emission color. The organic EL devices of the present invention accomplish improvements in light emission efficiency and light emission brightness and a longer device life, and does not impose any limitations on a light-emitting material, a dopant, a hole transporting material, an electron transporting material, a sensitizer, a resin and an electrode material used in combination and the method of producing the device. The organic EL device using the material of the present invention as a light-emitting material achieves light emission having a high brightness with a high light emission efficiency and a longer life as compared with conventional devices. According to the light-emitting material of the present invention and the organic EL device of the present invention, there can be achieved an organic EL device having a high brightness, a high light emission efficiency and a long life.

Claims

1. A pyridine compound of formula I

at least one of the groups W, X and Y is a group of formula

and the other groups are independently of each other an aryl group or a heteroaryl

group, especially a group of formula

 R^{11} , R^{12} , R^{12} , R^{12} , R^{13} , R^{13} , R^{15} , R^{15} , R^{16} , R^{16} , R^{17} , R^{17} , R^{41} , R^{41} , R^{42} , R^{42} , R^{44} , R^{44} , R^{45} , R^{45} , R^{46} , R^{46} , R^{47} and R^{47} are independently of each other H, E, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; C_7 - C_{18} aralkyl; or C_7 - C_{18} aralkyl which is substituted by E; or R^{11} and R^{12} , R^{12} and R^{13} , R^{15} and R^{16} , R^{16} and R^{17} , R^{44} and R^{46} and/or R^{45} and R^{47} are each a divalent group L^1 selected from an oxygen atom, an sulfur atom, $>CR^{18}R^{19}$

 R^{18} and R^{19} are independently of each other C_1 - C_{18} alkyl; C_1 - C_{18} alkoxy, C_6 - C_{18} aryl; C_7 - C_{18} aralkyl; or

 R^{11} and $R^{11'}$, R^{12} and $R^{12'}$, R^{13} and $R^{13'}$, $R^{13'}$ and R^{14} , R^{14} and R^{15} , R^{15} and $R^{16'}$, $R^{16'}$ and $R^{17'}$ and $R^{17'}$ and $R^{17'}$ and $R^{11'}$, R^{11} and $R^{11'}$, R^{12} and $R^{12'}$, $R^{12'}$ and $R^{13'}$, $R^{14'}$ and $R^{14'}$, R^{14} and $R^{14'}$, R^{15}

and R^{45} , R^{46} and R^{46} , R^{47} and R^{47} , R^{46} and R^{48} and/or R^{47} and R^{48} are each a divalent

group , wherei

 R^{30} , R^{31} , R^{32} , R^{33} , R^{49} and R^{50} are independently of each other H, C₁-C₁₈alkyl; C₁-C₁₈alkyl, which is substituted by E and/or interrupted by D; E; C₆-C₁₈aryl; C₆-C₁₈aryl, which is substituted by E;

 R^{14} is H, C_2 - C_{30} heteroaryl, C_6 - C_{30} aryl, or C_6 - C_{30} aryl which is substituted by E, C_1 - C_{18} alkyl which is substituted by E and/or interrupted by D; especially

 R^{21} , R^{22} , R^{23} , R^{24} , R^{25} , R^{26} and R^{27} are independently of each other H, E, C₁=C₁₈alkyl; C₁-C₁₈alkyl which is substituted by E and/or interrupted by D; E; C₇-C₁₈aralkyl; C₇-C₁₈aralkyl which is substituted by E;

 R^{43} and R^{48} are independently of each other H, E; C_1 - C_{18} alkyl; C_1 - C_{18} alkyl, which is substituted by E and/or interrupted by D; C_2 - C_{30} heteroaryl; C_7 - C_{18} aralkyl; C_7 - C_{18} aralkyl which is substituted by E;

D is -CO-; -COO-; -S-; -SO-; -SO₂-; -O-; -NR⁵-; SiR⁶¹R⁶²-; -POR⁵-; -CR⁶³=CR⁶⁴-; or -C=C-;

E is -OR⁵; -SR⁵; -NR⁵R⁶; -COR⁸; -COOR⁷; -CONR⁵R⁸; -CN; or halogen; wherein R⁵ and R⁶ are independently of each other C₆-C₁₈aryl; C₆-C₁₈aryl which is substituted by C₁-C₁₈alkyl, C₁-C₁₈alkyl; or C₁-C₁₈alkyl which is interrupted by -O-; or

R⁵ and R⁶ together form a five or six membered ring, in particular

 R^7 is C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by $-C_1$ -;

 R^8 is C_7 - C_{12} alkylaryl; C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by -O-;

 R^{61} and R^{62} are independently of each other C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by -O-, and R^{63} and R^{64} are independently of each other H, C_6 - C_{18} aryl; C_6 - C_{18} aryl which is substituted by C_1 - C_{18} alkyl, C_1 - C_{18} alkyl; or C_1 - C_{18} alkyl which is interrupted by -O- with

the proviso that compounds of formula I, wherein Y is

X and W are

; Y is

, X is

and W is

excluded.

2. A pyridine compound of formula I according to claim 1, wherein W, X and Y are independently of each other a group of formula

R¹¹, R¹², R¹², R¹³, R¹³, R¹⁵, R¹⁵, R¹⁶, R¹⁶, R¹⁶, R¹⁷ and R¹⁷ are independently of each other H, C₆-C₁₈aryl; C₆-C₁₈aryl which is substituted by E; E, C₁-C₁₈alkyl; C₁-C₁₈alkyl which is substituted by E and/or interrupted by D; C₇-C₁₈aralkyl; C₇-C₁₈aralkyl which is substituted by E; and

R¹⁴. R¹⁸ and R¹⁹ are as defined in claim 1, or

W is a group of the formula -W1-W2-W3,

X is a group of the formula -X1-X2-X3 and

Y is a group of the formula $-Y^1-Y^2-Y^3$, wherein W^1 , W^2 , X^1 , X^2 , Y^1 and Y^2 are independently of each other a group of formula

and W3, X3 and Y3 are independently of

$$- - R^{14}, - - R^{14}$$

each other a group of formula wherein R¹⁴ is as defined above.

- 3. The pyridine compound according to claim 1 or 2, wherein R¹¹, R¹¹, R¹², R¹², R¹³, R¹³, R¹⁵, R¹⁵, R¹⁶, R¹⁶, R¹⁷ and R¹⁷, R⁴¹, R⁴¹, R⁴², R⁴², R⁴⁴, R⁴⁴, R⁴⁵, R⁴⁵, R⁴⁶, R⁴⁶, R⁴⁷, and R⁴⁷ as well as R¹⁴, R⁴³, and R⁴⁸ are independently of each other H, E; or C₁-C₈alkyl; wherein E is -OR⁵; -SR⁵; -NR⁵R⁶; -COR⁸; -COR⁷; -CONR⁵R⁶; -CN; -OCOOR⁷; or F; wherein R⁵ and R⁶ are independently of each other C₆-C₁₂aryl, or C₁-C₈alkyl; R⁷ is C₇-C₁₂alkylaryl, or C₁-C₈alkyl; and R⁸ is C₆-C₁₂aryl; or C₁-C₈alkyl.
- 4. The pyridine compound according to any of claims 1 to 3, wherein

W, X and Y are a group of formula

wherein

 R^{13} , R^{15} and R^{15} are H and R^{20} is H, especially , or R^{13} and R^{15} are H, R^{13} and R^{15} are independently of each other H, C_1 - C_8 alkyl, or C_1 - C_8 alkoxy, and R^{20} is H, C_1 - C_8 alkyl, or C_1 - C_8 alkoxy; or

$$R^{32}$$
 R^{31} R^{33} R^{30} R^{30} R^{30} R^{30} R^{30} and R^{15} are H, and R^{13} and R^{20} are

$$R^{32}$$
 R^{31}
 R^{30}
 R^{30}
, wherein

 $R^{20},\,R^{15}\,\text{and}\,\,R^{15}$ are H, and $R^{13}\,\text{and}\,\,R^{13}\,\text{are}$

 R^{30} , R^{31} , R^{32} and R^{33} are H, C_1 - C_8 alkyl, or C_1 - C_8 alkoxy.

The pyridine compound according to any of claims 1 to 3, wherein
 W, X and Y are independently of each other a group of formula

$$R^{19} R^{18} R^{18}$$

$$R^{19} R^{19}$$

$$R^{19} R^{19}$$

$$R^{19} R^{19}$$

$$R^{19} R^{19}$$

$$R^{19} R^{19}$$

$$R^{19} R^{19}$$

$$R^{19$$

6. The pyridine compound according to claim 1, wherein
W and Y or W and X (= Y and X) are independently of each other a group of formula

$$R^{41}R^{41'} \qquad R^{44}R^{44'} \qquad R^{46}R^{46'} \qquad R^{48}$$
 is a group of formula
$$R^{42'}R^{42} \qquad , \qquad \text{or} \qquad R^{45}R^{45'} \qquad R^{47'}$$

wherein R^{11} , $R^{11'}$, R^{12} , $R^{12'}$, R^{13} , $R^{13'}$, R^{14} , R^{15} , R^{16} , R^{16} , $R^{16'}$, R^{17} , R^{41} , $R^{41'}$, R^{42} , $R^{42'}$, $R^{44'}$, R^{45} , $R^{46'}$, $R^{46'}$, R^{47} , R^{47} , R^{43} and R^{48} are defined as in claim 1, especially H, C_1 - C_8 alkoxy, or phenyl.

7. An electroluminescent device, comprising a pyridine compound of formula I according to any of claims 1 to 6 including compounds of formula I, wherein Y is

- 8. Electroluminescent device according to claim 7, wherein the electroluminescent device comprises in this order
 - (a) an anode
 - (b) a hole injecting layer and/or a hole transporting layer
 - (c) a light-emitting layer
 - (d) optionally an electron transporting layer and
 - (e) a cathode.
- 9. Electroluminescent device according to claim 8, wherein the pyridine compound of formula I forms the light-emitting layer.

10. Use of the pyridine compounds of formula I according to any of claims 1 to 6 for electrophotographic photoreceptors, photoelectric converters, solar cells, image sensors, dye lasers and electroluminescent devices.

Abstract

Disclosed are electroluminescent devices that comprise organic layers that contain pyridine compounds. The pyridine compounds are suitable components of blue-emitting, durable, organo-electroluminescent layers. The electroluminescent devices may be employed for full color display panels in, for example, mobile phones, televisions and personal computer screens.

This Page is Inserted by IFW Indexing and Scanning Operations and is not part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

□ BLACK BORDERS
□ IMAGE CUT OFF AT TOP, BOTTOM OR SIDES
□ FADED TEXT OR DRAWING
□ BLURRED OR ILLEGIBLE TEXT OR DRAWING
□ SKEWED/SLANTED IMAGES
□ COLOR OR BLACK AND WHITE PHOTOGRAPHS
□ GRAY SCALE DOCUMENTS
□ LINES OR MARKS ON ORIGINAL DOCUMENT
□ REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY

IMAGES ARE BEST AVAILABLE COPY.

As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.